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TITLE: Carbon product comprising carbonaceous materials joined together, said carbon product for electrode substrate of fuel cells and process for production thereof

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INVENTOR-INFORMATION:

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not both mixed

US-CL-CURRENT: 428/408; 428/137, 428/188, 429/44

CLAIMS:

What is claimed is:
1. A carbon product comprising carbonaceous materials and flexible graphite sheets interposed between said carbonaceous materials, wherein (1) said carbonaceous materials and said flexible graphite sheet are joined together, (2) the thus joined materials have been integrated by calcination as a unitary carbon body in an inert atmosphere, (3) the joining surface of at least one of said carbonaceous materials comprises both joining parts and non-joining parts which have an optional shape and are uniformly arranged on the joining surface, (4) the ratio of the total area of said non-joining parts in said joining surface of said carbonaceous material to the total area of said joining surface is from 0.20 to 0.80, and (5) the value of the difference of the rate (%) of linear expansion and contraction at calcination between two said carbonaceous materials to be subjected to mutual joining via said flexible graphite sheet is not more than 3%.
2. A carbon product according to claim 1, wherein said carbonaceous material having said non-joining parts is a plate having depressed parts.
3. A carbon product according to claim 1, wherein the parts of said flexible graphite sheet, corresponding to said non-joining parts of said carbonaceous material, have been removed.
4. A carbon product according to claim 1, wherein the ratio of the total area of said non-joining parts on said joining surface of said carbonaceous material to the total area of said joining surface is from 0.30 to 0.70.
5. A carbon product according to claim 1, wherein said carbon product has been calcined as a whole at a temperature of now lower than 800.degree. C. in an inert atmosphere.
6. A carbon product according to claim 1, wherein said carbonaceous materials are selected from the group consisting of
(1) molded carbonaceous materials comprising a carbonaceous aggregate and a binder, optionally and an organic granular substance,
(2) carbonaceous materials obtained by calcining molded materials of the above (1) in an inert atmosphere,
(3) molded carbonaceous materials comprising a graphitic aggregate and a binder, and
(4) carbonaceous materials obtained by calcining molded materials of the above (3) in an inert atmosphere.
7. A carbon product according to claim 6, wherein said carbonaceous aggregate is at least one kind of aggregate selected from the group consisting of carbon

fibers, carbon particles and oxidized pitch particles.
8. A carbon product according to claim 6, wherein said binder is at least one kind of binder selected from the group consisting of phenol resins, furan resins, epoxy resins, petroleum pitches and coal pitches.
9. A carbon product according to claim 6, wherein said graphitic aggregate is graphite particles, easily graphitizabIe carbonaceous particles or both.
10. A carbon product according to claim 1, wherein said flexible graphite sheet is prepared by compressing expanded graphite particles obtained by subjecting graphitic particles of not more than 5 mm in diameter to acid-treatment and further to heating treatment and have a thickness of not more than 1 mm, a bulk density of 0.5 to 1.5 g/cm.sup.3 and a coefficient of compression strain of not less than 10.sup.-4 cm.sup.2 /kg.
11. A carbon product according to claim 1, wherein said carbonaceous material having non-joining parts of the joining surface is an elecrode part material comprising a porous carbonaceous flat plate which has a plurality of carbonaceous protuberances uniformly arranged on the joining surfaces thereof, and said carbonaceous protuberances have been joined to the other of said carbonaceoss materials interposing said flexible graphite sheet therebetween.
12. A unitary electrode substrate for fuel cells, which substrate comprises (1) a carbonaceous material for a separator which bulk density is not less than 1.2 g/cm.sup.3, (2) flexible graphite sheets placed on each of and joined to the two surfaces of said carbonaceous material for a separator, and (3) a plurality of carbonaceous protuberances on outer surfaces of said flexible graphite sheets and a pair of porous carbonaceous flat plates on outermost surfaces of the electrode substrate as an electrode materials, wherein all of the materials of said electrode substrate is integrated as a unitary carbon body by calcination in an inert atmosphere, and the value of the difference of linear expansion and contraction rates at calcination between the carbonaceous material for the electrode material and the carbonaceous material for the separator is not more than 3%, and the ratio (Sr/Se) of the total area of cross section of said carbonaceous protuberances, which cross section is parallel to a surface of said carbonaceous material for said separator (Sr), to the total area of the joining surface of said porous carbonaceous flat plate (Se) is from 0.2 to 0.8.
13. An electrode substrate according to claim 12, wherein the space between the neighboring protuberances is not more than 10 mm.
14. An electrode substrate according to claim 12, wherein said carbonaceous protuberances are arranged in series.
15. An electrode substrate for fuel cells according to claim 12, wherein said carbonaceous protuberances are arranged in an alternating pattern.
16. An electrode substrate according to claim 12, wherein said porous carbonaceous flat plates have an average bulk density of 0.25 to 0.9 g/cm.sup.3 and a gas-permeability of not less than 30 cm.sup.2 /hour mmAq., and an average bulk density of said carbonaceous protuberances is 0.40 to 1.8 g/cm.sup.3.
17. A unitary electrode substrate for fuel cells, which substrate comprises:
(1) a carbonaceous material for a separator which bulk density is not less than 1.2 g/clm.sup.3,
(2) a flexible graphite sheets placed on each of and joined to the two surfaces of said carbonaceous material for a separator.
(3) a plurality of carbonaceous protuberances on outer surfaces of said flexible graphite sheets and a pair of porous carbonaceous flat plates on outermost surfaces of the electrode substrate as an electrode material, wherein (i) all of the materials of said electrode substrate is integrated as a unitary carbon body by calcination in an inert atmosphere, and (ii) a plurality of passages for gaseous reactants are defined by (a) a flexible graphite sheet or the combination of a flexible graphite sheet and the carbonaceous separator, (b) the plurality of carbonaceous protuberances, and (c) the porous carbonaceous flat plate, (iii) the carbonaceous protuberances are arranged for uniform gaseous reactant distribution within the passages formed by the protuberances, (iv) the ratio (Sr/Se) of the total area of cross section of said carbonaceous protuberances, which cross section is parallel to a surface of said carbonaceous material for said separator (SR), to the total area of the joining surface of said porous carbonaceous flat plate (Se) is from 0.2 to 0.8, and (v) the value of the difference of linear expansion and contraction rates at calcination between the carbonaceous material for the electrode material and the carbonaceous material for the separator is not more than 3%.